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ELECTROCHEMICAL TREATMENT OF MUNICIPAL WASTE WATER

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ELECTROCHEMICAL TREATMENT OF MUNICIPAL WASTE WATER

by

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for

The Advanced Waste Treatment Research Program
Basic and Applied Sciences Branch
Robert A. Taft Sanitary Engineering Center

A report submitted in fulfillment of a contract between the Public Health Service and Pennsalt Chemicals Corporation (Contract No. PH 86-62-113). Submitted October 1963.

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

Division of Water Supply and Pollution Control

March 1965

The ENVIRONMENTAL HEALTH SERIES of reports was established to report the results of scientific and engineering studies of man's environment: The community, whether urban, suburban, or rural, where he lives, works, and plays; the air, water, and earth he uses and re-uses; and the wastes he produces and must dispose of in a way that preserves these natural resources. This SERIES of reports provides for professional users a central source of information on the intramural research activities of Divisions and Centers within the Public Health Service, and on their cooperative activities with State and local agencies, research institutions, and industrial organizations. The general subject area of each report is indicated by the two letters that appear in the publication number; the indicators are

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Public Health Service Publication No. 999-WP-19

ADVANCED WASTE TREATMENT

RESEARCH PROGRAM

The Advanced Waste Treatment Research Program of the Public Health Service has two ultimate goals: One is to help abate our Nation's growing water pollution problems, and the other, more startling in concept, is to renovate waste water for direct and deliberate re-use.

Conventional water and waste treatment practices have little or no effect on many simple inorganic salts and permit an ever-increasing number and amount of highly complex, synthetic organic wastes to contaminate drinking water supplies. If we are to protect these water supplies for the future, it is mandatory that new technologies for water and waste treatment be developed.

Advanced Waste Treatment may be looked upon as a two-step process: (1) Separating concentrated contaminants from the purified water "product," and (2) disposing of the concentrated contaminants in a way that will render them forever innocuous. This "permanent disposal" of separated wastes is essential so that useable surface or ground waters will not be contaminated and so that the same contaminants need not be removed again and again from water supplies.

In one step, then, advanced waste treatment, i.e., water renovation, could alleviate both water pollution and water supply problems -- problems of increasing concern both in this country and in the rest of the world. The importance of advanced waste treatment was recognized by Congress in its 1961 amendments to the Federal Water Pollution Control Act (Public Law 87-88). This act directs the Secretary of Health, Education, and Welfare ". . . to develop and demonstrate practicable means of treating municipal sewage and other water-borne wastes to remove the maximum possible amounts of physical, chemical, and biological pollutants in order to restore and maintain the maximum amount of the Nation's water at a quality suitable for re-use.

It is too early to predict accurately the cost of advanced waste treatment technology, but the need is inevitable. Because of population increase and industrial expansion, we are approaching the time when our rivers and streams will no longer be able to assimilate our wastes and when there will be no more developable supplies of fresh water. Whatever the cost of this technology, one thing is certain -- it must be the very lowest science

and engineering can achieve. Anything more will impose an inexcusable, perhaps intolerable burden on our country's economic growth and on its ability to compete in tomorrow's world.

The Pennsalt Chemicals Corporation has broad experience in electrochemistry and electrochemical processing techniques. For over 100 years, Pennsalt has been closely associated with the electrochemical industry, first through the production of chlorine and caustic soda and later through large-scale production of such diverse materials as chlorates, perchlorates, hydrogen peroxide, persulfates, and elemental fluorine.

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ABSTRACT

Although electrolytic methods of treating waste water are reported in the older literature, their effectiveness is in doubt. This study has shown that an electrolytic treatment of secondary effluent from an activated-sludge plant using lead dioxide anodes can reduce ABS by 90 percent, COD by 80 percent, and chloride by 90 percent. This treatment has little effect on total solids. The cost of treatment will vary between $0.90 and $2.42 per 1,000 gallons. The plant costs will vary between $9 million and $26 million for a 10 mgd plant. These costs do not include site preparation, roads, landscaping, fences, land area, lighting, laboratories, or other buildings, and the costs depend on the completeness of the treatment and the electrical conductivity of the waste water. The method is not considered economically practical for waste-water renovation, wherein waters of relatively low conductivity must be processed.

ELECTROCHEMICAL TREATMENT OF MUNICIPAL WASTE WATER

Objective of This Study

The objective of this study was to investigate the possibility of using an electrolytic method for the removal of refractory contaminants from the effluent of municipal waste-water treatment plants.

Summary

1. A literature survey was made of electrolytic methods of treating waste water.

A considerable amount of work was done on electrolytic waste-water treatment from 1900 to 1930. This work resulted in the establishment of several electrolytic municipal waste plants, but all were eventually abandoned; the reasons given at the time were high cost of operation and questionable efficiency. Waste water mixed with sea water is now being electrolyzed at two European plants. Satisfactory separation of solids at a cost of about 6 cents per 1,000 gallons of waste is claimed at a power cost of 1 cent per kilowatt-hour. No work on the electrolytic treatment of municipal waste has been reported in the United States in recent years.

2. A procedure was set up to evaluate electrolytic methods of treating samples from the Abington Sewage Plant, Abington, Pennsylvania. Standard analytical procedures for COD, ABS, chlorides, and total solids were recommended or supplied by the Robert A. Taft Sanitary Engineering Center.

3. The specific conductance was determined for both primary and secondary effluents from the Abington plant.

4. The performance of various electrode materials was evaluated for use in the electrolysis of waste water. Erosion rates and polarization potentials were determined in both secondary effluent and KCl solutions having the same conductance.

5. The standard analytical procedures were modified to work on samples containing hypochlorite. A faster method for the determination of ABS was demonstrated.

6. An apparatus for the pasteurization of secondary effluent by a procedure devised by the Advanced Waste Treatment Research Program was set up.

7. A series of statistically designed experiments was run with selected anode materials to determine optimum conditions of pH, current density, and temperature for the electrolytic treatment of pasteurized secondary effluent.

8. The destruction of bacteria by electrolysis was determined for one electrode system.

9. The effect of sodium hypochlorite and chlorine on the reduction of COD, ABS, and total solids in secondary effluent was determined.

10. Two sizes of cells were run at optimum conditions to yield data on the power requirement and performance of the lead dioxide anode-iron cathode cell.

11. The data were analyzed and estimates made on the cost of the most promising electrolytic process for the treatment of secondary effluent.

Conclusions

1. The early electrolytic treatment using iron electrodes in a waste solution treated with lime was probably effective because of the relatively high pH and small amount of hypochlorite formed. There is no evidence that any organic material was decomposed by the electrolytic action.

2. If reasonable power is to be used, the very low electrical conductivity of the primary and secondary effluent requires very close spacing between the anode and cathode of a cell to electrolyze such solutions. This low conductivity eliminates the possible use of a diaphragm-type cell.

3. Electrolytic action may prove a very efficient way to introduce chemicals into waste water. An eroding iron anode can introduce an iron floc uniformly and at a constant rate without the introduction of an anion. A lead dioxide anode can convert chloride ion in a waste solution to hypochlorite ion at a uniform and controlled rate without introducing any new materials into the solution.

4. Iron, lead, magnesium, aluminum, graphite, and copper all erode when made anodic in secondary effluent, and can be used only under special conditions.

5. Lead dioxide, platinum, and platinum-plated titanium do not erode when made anodic in secondary effluent.

6. All electrode materials except lead dioxide worked satisfactorily as the cathode in secondary effluent.

7. Graphite, lead dioxide, platinum, and platinum-plated anodes generate significant amounts of hypochlorite when used in secondary effluent.

8. Hypochlorite ion interferes with chloride, ABS, and COD determinations. It can be destroyed by H_2O_2 for the ABS determination and by titanous sulfate in the COD analysis.

9. Lead dioxide was shown to be the most effective anode material for the reduction of ABS, COD, and chloride in waste water. It did not reduce total solids, suggesting that the treatment did not decompose the dissolved material into volatile material.

10. Electrolysis with a "noble" inert anode is very effective in the destruction of bacteria.

11. The presence of hypochlorite ion in electrolyzed waste water does not account for the reduction of ABS or COD by the electrolytic treatment.

12. The electrolysis of secondary effluent with lead dioxide anodes and platinum-plated titanium cathodes can reduce the COD by as much as 80 percent, ABS by as much as 90 percent, and chloride by as much as 90 percent. Lead dioxide anodes with iron cathodes may be somewhat less efficient in reducing COD. All systems have little effect on total solids.

13. The cost of treating secondary effluent by the electrolytic method will vary between $0.90 and $2.42 per 1,000 gallons depending on the completeness of the treatment and the electrical conductivity of the waste water. The plant costs, even with very optimistic assumptions, will vary between $9 million and $26 million, for a 10 mgd plant.

Recommendations

1. Because of its comparatively high cost, the electrochemical treatment of waste water cannot be recommended as a process for the Advanced Waste Treatment Research Program, where waters of relatively low conductivity must be processed.

2. The work by the Lead Industries Association now underway on the use of lead and lead-silver alloys in the treatment of waste waters should be followed. Their results may have application in treating waste waters of relatively high conductivity.

Report

INTRODUCTION

The purpose of this study was to determine the feasibility of using an electrolytic process to remove, destroy, or render less objectionable the trace dissolved material in waste-water effluent, so that this waste water could be re-used.

The effort was not directed towards the development of a system to replace present methods of waste-water treatment, but was intended to develop a satisfactory posttreatment process for removal of so-called refractory compounds not removed by conventional waste-water treatment. Although many of these refractory compounds are known, there are many that are not known. It is not possible, therefore, to make synthetic waste-water samples. In this study, at the suggestion of the Project Officer at the Taft Sanitary Engineering Center, liquid samples were taken from primary and secondary waste-water treatment plant streams. The scope of work required that the effect of the waste-water treatment be measured in terms of the standard waste-water analytical procedures of chemical oxygen demand (COD), total solids (TS), chloride ion (Cl), and alkylbenzenesulfonate (ABS). The criterion for satisfactory performance of an electrode system was the degree of reduction of the four quantities, COD, ABS, TS, and chlorides.

Because this project was exploratory, it was planned that the experimental approach should be to use as much intuition and insight as possible so as to move quickly to those techniques and procedures that give most promising results. Effort was directed towards determining the feasibility of the electrolytic method rather than towards generating technical or scientific data.

LITERATURE REVIEW

During the first quarter of this century, electrolytic waste-water treatment enjoyed a considerable vogue. The most publicized method was that of C. P. Landreth. Essentially, this method treated raw waste water with lime and electrolyzed it with iron electrodes. The solids were allowed to settle and the effluent was discharged. The effluent was described as being clear, odorless, and nonputrescent. Several demonstration plants were built and became the center of considerable controversy. Doubt was expressed in the literature about the effectiveness of the method; in fact, some critics questioned whether it worked at all. It was claimed that the lime treatment and settling produced the same result whether or not the waste water was electrolyzed.

Estimates for relatively large systems indicated that the electrolytic method would cost about double that of methods then in use. This, it is felt, was the real reason the method failed to gain more acceptance. The nonelectrolytic methods then in use were, however, relatively simple, and when judged against the cost of a modern activated-sludge plant, this electrolytic method would be competitive costwise. Data on its actual performance do, however, leave some question about its effectiveness by present standards.

Several electrolytic methods for the treatment of industrial wastes have appeared in the patent literature since the twenties, but very little has been done on a large scale. Recently, however, interest has been revived in these methods. Two groups, one in Italy and another in Norway, are studying methods using sea-water addition to increase the conductivity. The mixed waste water and sea water is electrolyzed and then discharged into the ocean. The methods show promise but are not feasible for use where the effluent cannot be discharged directly into the sea.

Appendix 1 is a discussion of electrolytic methods and Appendix 2 is a bibliography of the literature on electrolytic waste-water treatment.

EXPERIMENTAL WORK

Abington Township Sewage Plant

All waste-water samples for this study were obtained from the waste-water treatment plant of Abington Township, Montgomery County, Pennsylvania. This source was approved by the Project

Officer at the Taft Sanitary Engineering Center. The plant serves a population of about 26,000 and receives an average of 2.4 million gallons per day (mgd) of waste water. This is almost entirely sanitary waste. A flow diagram is shown in Figure 1. The plant uses an activated-sludge process.

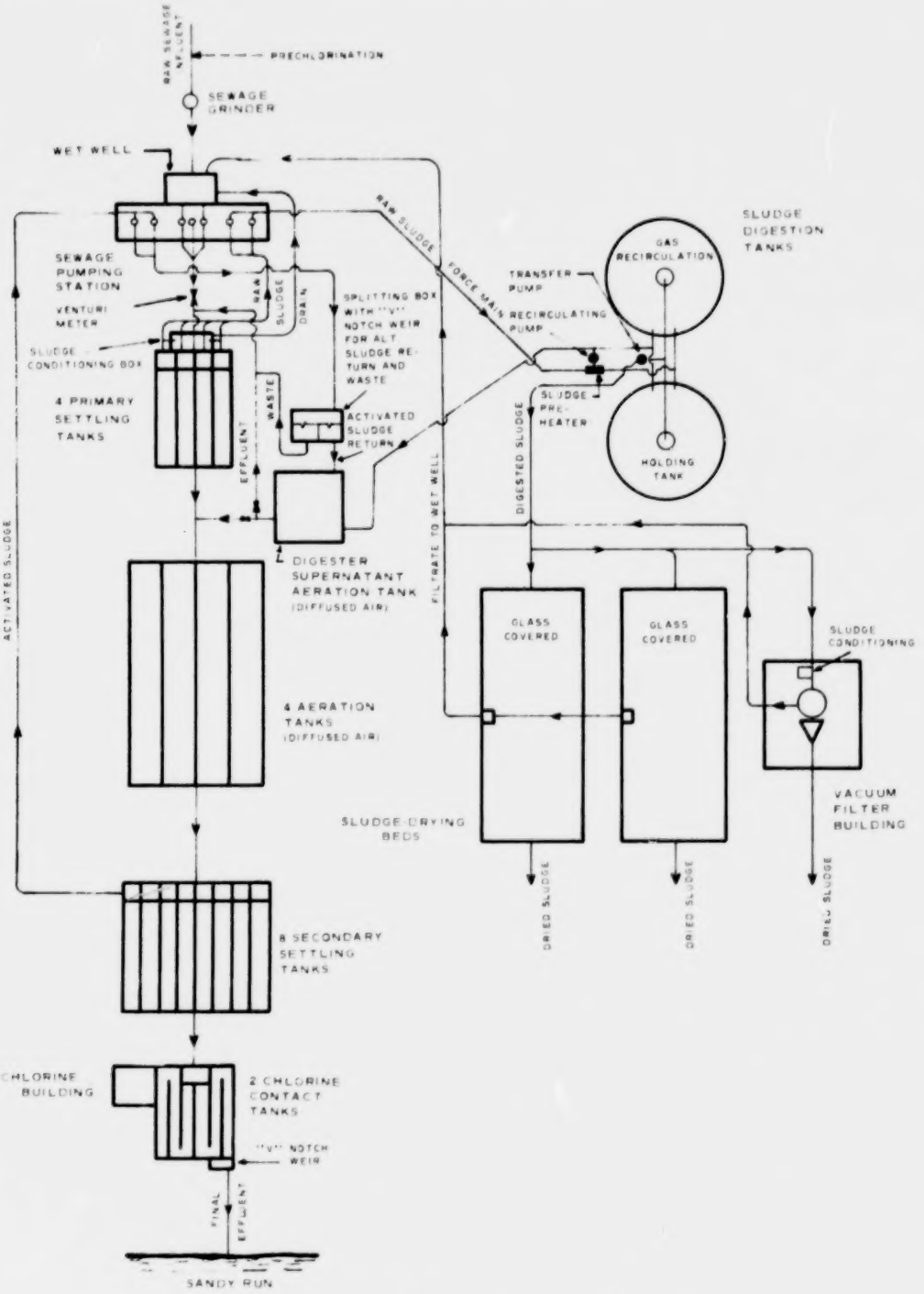

Figure 1. Flow diagram. Sewage Treatment Plant, Abington Township, Montgomery County, Pa.

Raw waste water is passed through a primary settling tank, an aeration tank, and a secondary settling tank. The effluent from the secondary tank is chlorinated and discarded. Waste activated sludge from the secondary settling tanks is returned to the primary tanks. Sludge from the primaries is taken to two sludge digestion tanks, 75 feet in diameter and 32 feet high. They are operated in series, and anaerobic digestion is carried on at temperatures controlled between 95 and 100° F. Sludge is sent to the tanks daily, and the holdup time for digestion is about 14 days.

Considerable volumes of gas are formed in the digestion tanks. Some of the gas is recirculated to the bottom of the tanks for agitation and the rest is bled off through a water trap and used for heating or is flared.

The supernatant liquid from the sludge digestion tank is returned to the raw waste-water inlet. Part of the sludge from the digestion tanks goes to drying beds enclosed in greenhouse-like structures where drying proceeds for about 6 days. The rest of the sludge goes to a vacuum filter where it is dosed with 5 percent ferric chloride and 14 percent lime on the basis of the dry solids. The filtrate is piped back to the raw waste-water inlet.

The raw waste water has a BOD of about 260 parts per million (ppm) and suspended solids of about 330 ppm. Effluent from the primary tanks has a BOD of about 200 ppm and suspended solids of 250 ppm. Most of the samples for this work were taken from the secondary-effluent stream before final chlorination. The BOD at this point is about 40 ppm and suspended solids, 30 ppm. A series of 24-hour composites was analyzed, and average values obtained, in ppm, were: COD, 60; total solids, 475; chloride, 50; and ABS, 8. All samples regardless of source were filtered through Whatman No. 5 filter paper before use. Specific conductance was in the range 5.5 to 6.2 x 10^{-4} mho per centimeter for primary effluent and 4.75 to 5.3 x 10^{-4} for secondary effluent. Analyses of samples of primary and secondary effluent are shown in Table 1.

Apparatus and Procedures

All the electrolyses carried out in this study used direct current obtained either from lead storage batteries or from a line-operated power supply that delivered direct current with only a negligible amount of ripple. Voltage and current to the cells were measured with Weston precision meters.

Table 1. ANALYSIS OF ABINGTON TOWNSHIP FILTERED EFFLUENT

Date	Type effluent	Specific conductance, mhos/cm x 10^4	COD, ppm	Total solids,[a] ppm	Chloride, ppm	ABS, ppm	pH
5/9/62	Primary	6.09	114	420	46	---	8.1
5/9/62	Secondary	5.19	41	460	48	---	7.5
5/10/62	Primary	5.95	98	450	52	---	8.2
5/10/62	Secondary	5.32	38	490	53	---	7.35
5/11/62	Primary	5.68	82	460	46	---	8.15
5/11/62	Secondary	5.05	47	500	43	---	7.5
5/14/62	Primary	5.82	103	430	42	---	8.1
5/14/62	Secondary	5.05	50	500	48	---	7.45
5/15/62	Primary	6.11	102	450	49	---	8.05
5/15/62	Secondary	5.25	53	540	52	---	7.55
5/16/62	Primary	6.05	101	490	49	---	8.1
5/16/62	Secondary	5.25	64	520	46	---	7.6
5/17/62	Primary	5.75	95	410	50	---	8.15
5/17/62	Secondary	5.19	66	520	52	---	7.7
5/18/62	Primary	5.98	102	440	52	---	8.1
5/18/62	Secondary	5.24	58	470	52	---	7.6
5/21/62	Primary	5.62	103	420	47	---	8.2
5/21/62	Secondary	4.95	62	490	48	---	7.6
5/22/62	Primary	6.30	149	520	55	---	8.1
5/22/62	Secondary	5.28	71	490	50	---	7.9
5/23/62	Secondary	5.56	82	490	52	---	7.8
5/31/62	Secondary	5.40	74	470	51	---	7.5
6/12/62	Secondary	5.05	55	490	50	---	7.2
6/14/62	Secondary	4.51	51	450	42	---	7.3
6/18/62	Secondary	5.07	50	450	46	6.9	7.2
6/19/62	Secondary	5.50	60	460	52	8.7	7.65
6/25/62	Secondary	5.16	45	490	49	6.1	7.4

[a] These data are for total solids on filtered effluent and represent total dissolved solids. Total solids in unfiltered effluent were not determined in this study, but run about 680 ppm for primary and 520 ppm for secondary effluent for this plant.

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Several types of electrolytic cells were used. In all cases the materials used in their construction were chosen so as not to contribute to the COD of the test solutions. All cells were checked for this possibility by filling them with distilled water and allowing them to stand overnight. In no cases were significant amounts of COD found in the distilled water. Polyethylene, glass, and "Stop-Off," * a lacquer material, were used as insulation in the cells.

Figure 2 shows a typical setup used in the anode screening tests. The anodes and cathodes alternated in the electrode pack. The active dimension of each electrode was 3 by 3 inches. There were five anodes and six cathodes. Later runs used a system containing only three electrodes, two cathodes, and one anode. These were held about 1/4 inch apart by polyethylene supports. Systems of more than 1-liter volume utilized a circulation system driven by a Gorman-Rupp pump constructed by Buna-N rubber and stainless steel. Electrolyte was withdrawn from the bottom of the cell and returned through a showerhead. This gave an effective distribution over the electrodes and aided in reducing foam levels. Studies using less than a liter of solution were carried out in a beaker stirred with a Teflon-covered stirring bar. Figure 3 is a typical setup.

All pH measurements were done with a Leeds and Northrup line-operated laboratory meter. Conductivity measurements were done with a Leeds and Northrup 60-cycle conductivity bridge No. 4688. All chemicals used were CP grade unless otherwise stated.

The anode screening test used grab samples of raw waste water from the Abington plant. All other tests, unless therein stated, used pasteurized secondary effluent from the Abington plant prepared by a procedure supplied by the Taft Sanitary Engineering Center. Pasteurized secondary effluent has been observed for over 3 weeks with little change in its analysis. Analyses of five typical batches are shown in Table 2.

Although it has been reported by the Taft Center that this method often precipitates some of the phosphate, we have not had this problem. In fact, no precipitate of any kind has been observed during storage, and only one sample gave an unusual analysis after pasteurizing and storage.

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* A product of Michigan Chrome and Chemical Company.

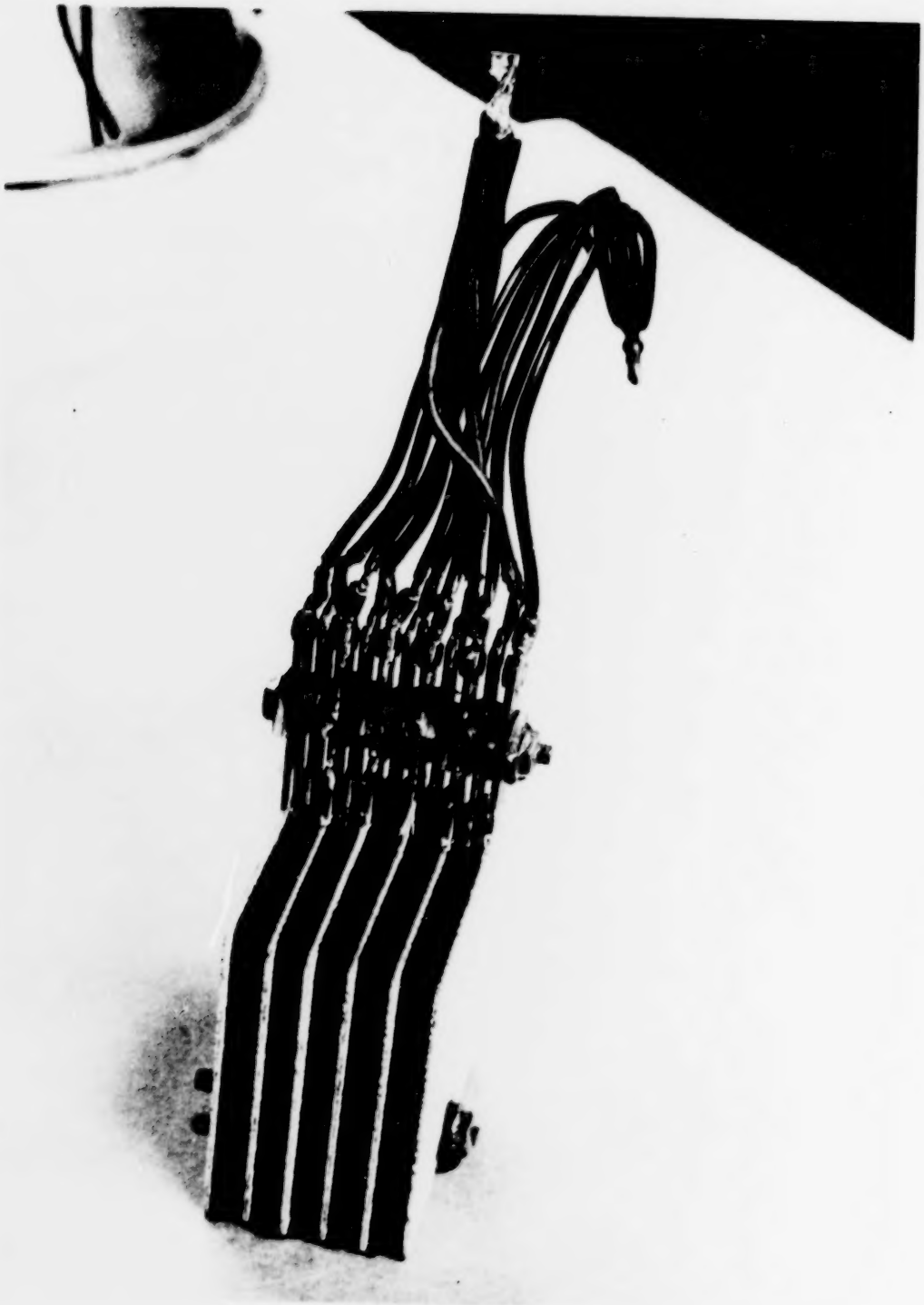

Figure 2. Grid of nine electrodes.

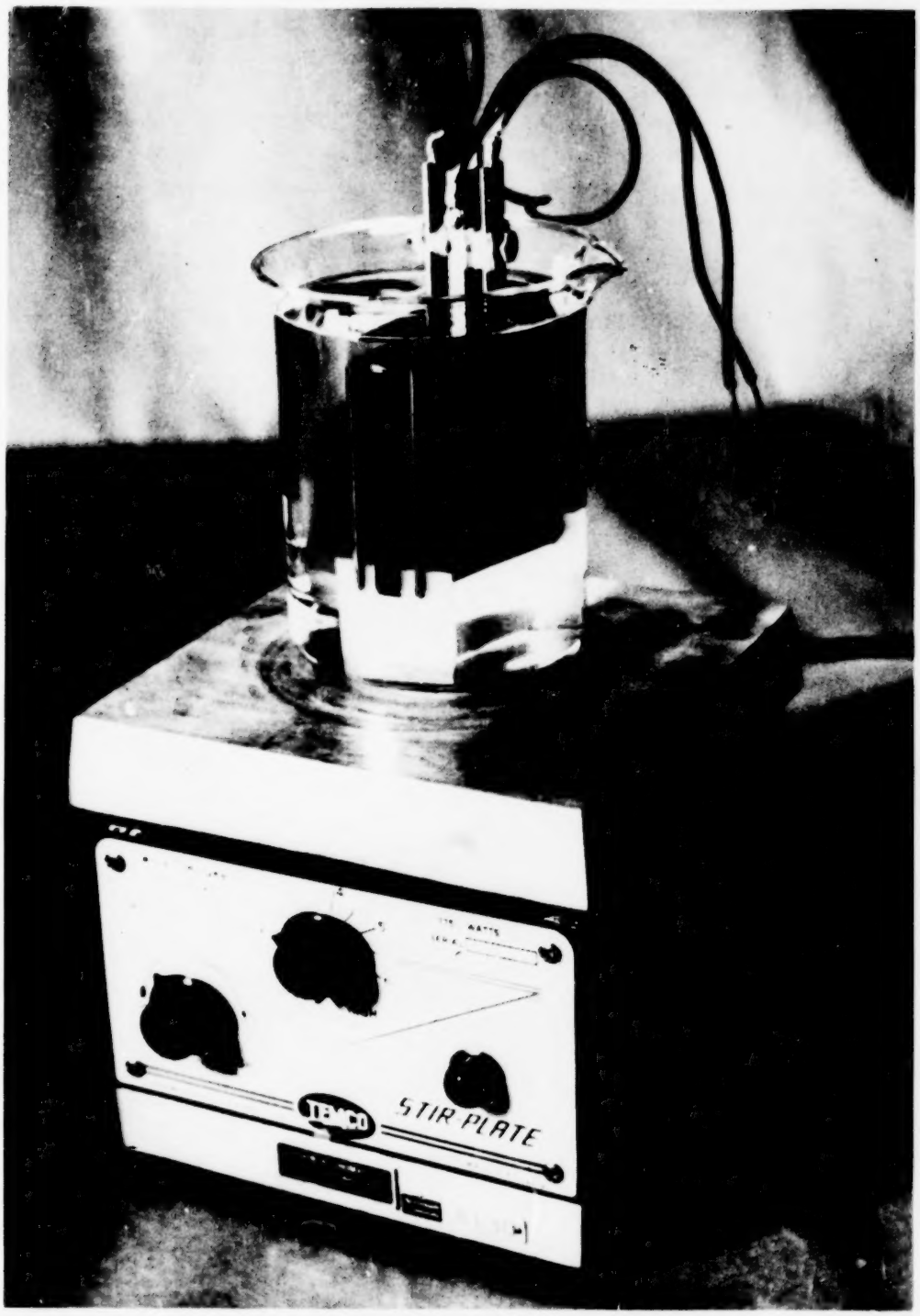

Figure 3. Cell with magnetic stirring.

At the start of the project all analytical methods used were standard, either AWWA or Taft Center methods. The analytical determinations made were the Taft Center's methylene blue method for determining alkylbenzenesulfonate (ABS), and AWWA methods for chemical oxygen demand (COD), total dissolved solids (TS), and chlorides (Cl). These methods were used exactly as written, with no corrections or changes for any extraneous materials in the samples.

Table 2. PASTEURIZED SECONDARY EFFLUENT AGING DATA

Date of sample	COD, ppm	TS, ppm	Cl, ppm	ABS, ppm	Before or after pasteurizing
Batch I					
12/11/62	48	570	70	7	After
12/18/62	45	560	71	7	"
12/26/62	50	566	72	7	"
Batch II					
12/31/62	49	491	75	8	Before
1/2/63	57	490	72	9	After
1/7/63	57	491	66	8	"
1/14/63	67	630	88	8	"
Batch III					
1/21/63	59	460	58	6	Before
1/24/63	59	470	57	7	After
2/4/63	58	460	56	6	"
Batch IV					
2/13/63	46	420	53	7	Before
2/14/63	47	420	55	7	After
2/20/63	49	380	54	6	"
2/27/63	48	410	49	8	"
3/7/63	42	400	54	6	"
3/20/63	39	390	52	7	"
Batch V					
3/18/63	43	390	49	7	Before
3/20/63	50	390	47	8	After
3/27/63	37	390	50	8	"

During the 5th month of the project, a Hellige comparator for determining ABS in water was obtained. This instrument was checked against standard samples and against the usual standard method and found to give results within the limits of error of the methods. It was then used in place of the Taft Center method since it was much faster.

During the early screening tests, it became evident that hypochlorite was being formed in the electrolyzed solution. Investigation showed that this hypochlorite (available chlorine) interfered in the determination of total chloride, COD, and ABS. Methods overcoming these interferences were worked out and used from then on. A method for the analysis of available chlorine was also worked out.

Because of the color of the samples treated with graphite anodes, the end point in the Mohr method for the determination of total chloride could not be seen. An amperometric titration apparatus was set up to determine chlorides on these samples.

The presence of chlorate was suspected in some of the electrolyzed material, and a method was developed to determine chlorate content. Time did not, however, permit a full evaluation of this method.

Three samples were analyzed for: Total nitrogen, amino nitrogen, nitrate nitrogen, phosphate, sulfate, calcium, sodium, heavy metals, and loss on heating at 600° C. Methods used were either AWWA or adapted standard methods. Except for the chlorate method, all new methods were tested fully against standard samples; analyzed, unknown samples; and prepared, synthetic, unknown samples. The method was not adopted unless results agreed within the limit of error of the analyses.

Electrolyte Conductivity and Cell Design

The measured specific conductance of the secondary effluent from the Abington Township plant was found to range from 4.7×10^{-4} to 5.3×10^{-4} mho per centimeter. This is about the same conductivity as 0.003 molar KCl solution. This low conductivity places serious restrictions on the electrolytic processes to be used if power consumption is to be kept within reasonable limits.

Table 3 lists the voltage of an electrolytic cell using secondary effluent and brackish water as electrolyte at several current densities with electrodes spaced about 0.2 inch apart. The high voltages at the higher current densities are caused in most part by the voltage drop across the electrolyte. For example, the decomposition voltage of the electrolyte plus normal hydrogen and oxygen overvoltage at 30 milliamperes per square centimeter is estimated at less than 3.5 volts of the 27.9 volts measured. The remaining 24.4 volts is caused by the resistance drop of the electrolyte. The lower voltage shown in Table 3 for brackish-water electrolyte illustrates the advantage of using highly conductive solution.

It was found that electrode spacing of 1/8 to 1/4 inch was sufficient to allow good circulation of solution between electrodes and close enough to keep power requirements within reasonable levels at the lower current densities.

Table 3. CELL VOLTAGE VERSUS CURRENT DENSITY FOR TWO TYPES OF ELECTROLYTE

Current density, ma/cm^2	Cell voltage	
	Secondary effluent	Brackish water[a]
1	3.3	3.2
6	7.8	5.5
30	27.9	14.9

[a] Estimated voltage for a specific conductance of 12×10^{-4} mho/cm at a salt concentration of 1,000 ppm.

Cells with diaphragms were found to operate at voltages three times higher than those without diaphragms even at very close spacing of the electrodes to the diaphragm. No further work was done with this type cell because of the excessive power requirements.

Anode Material Screening

It is well known that the electrode material can influence the course of the electrolytic reaction. In this study, therefore, electrode materials were chosen that were known to differ in their electrolytic action. Anode materials were considered before cathode materials because of the greater probability of decomposing the organic waste by an oxidation reaction and because of the greater dependence of anodic reaction on the electrode material.

Noble anodes, not attacked during electrolysis, are the type of greatest interest to this study. The second type, eroding anodes, have been used in waste treatment, and in some cases the eroded metal formed a floc that helped to remove material from solution. In most cases, however, an eroding anode would be too expensive for waste treatment. In neutral or acid anolyte, platinum, magnetite, graphite, and lead dioxide are likely inert anode materials. In strong alkaline solution, iron and stainless steel should also be inert. Copper, lead, magnesium, and aluminum are of possible interest as eroding electrodes.

In the preliminary tests, all the metals were made up into grids consisting of five anodes and six cathodes as shown in Figure 2. The platinum anodes were made by J. Bishop Platinum Works and consisted of 100 microinches of platinum electroplate on a titanium base. This type of electrode is known to have a low chlorine overvoltage and a comparatively long life in a salt brine electrolysis. The lead dioxide anodes were prepared in this laboratory and consisted of a thick electroplate of dense lead dioxide on a tantalum screen. Lead dioxide anodes are used to make perchlorates by the electrolysis of a chlorate solution and are known to have a comparatively high chlorine overvoltage. In the preliminary tests, lead dioxide, carbon, and graphite were run in the screening tests in a cell using a single anode with two cathodes as shown in Figure 3.

Table 4 shows the changes in pH, COD, ABS, TS, and chlorides effected by each of the materials as tested. Current densities for all runs were approximately 6 milliamperes per square centimeter. The steel, copper, lead, magnesium, and aluminum dissolved almost quantitatively with current. The stainless steel was not attacked to as great an extent, but some precipitate was formed. It is probable that most of the effect shown on COD and ABS for these materials was due to absorption of impurities on the hydrated oxide precipitates. The various types of graphite were all attacked; rather than form a precipitate, they flaked, leaving fair amounts of particles in suspension. The solutions ranged in color from light brown to black. By far the most satisfactory materials were lead dioxide and platinum-plated titanium. On the basis of these results it was decided to limit further investigation to platinum-plated titanium, lead dioxide, and graphite. Graphite was included because it is commonly used in commercial electrolytic processes and it was hoped conditions would be found where it would not be attacked.

The attack on steel, steel alloys, magnesium, aluminum, and graphite was in agreement with published information. The lead-silver alloy is reported to resist attack in sea water at a current density of about 6 milliamperes per square centimeter. Under these conditions, a lead dioxide surface that resists further attack is reported to form. Our preliminary tests did not show this behavior, and since another type of lead dioxide anode was available, no further work was done with the lead-silver alloy.

Polarization Studies

Polarization measurements were performed on the materials of prime interest to determine any unusual effects they might

Table 4. SUMMARY OF ANODE SCREENING TESTS

Anode material	Time, hr	pH		COD, % change	Total solids, % change	Chlorides, % change	ABS, % change	Remarks
		Before	After					
430 Stainless steel	2	7.2	8.3	-29	-22	-8	-40	Seriously attacked
	5	7.6	9.5	-12	-17	-8	-44	
Copper	2	7.3	9.2	-39	-38	-2	0	Seriously attacked
"	2	7.2	9.5	-32	-33	-9	-29	
"	5	7.4	10.6	+100	-47	-80	-26	"
Aluminum	2	7.2	9.5	-8	-44	-7	-50	Seriously attacked
"	5	7.3	9.4	+25	-43	0	-63	
Magnesium	2	7.1	11.1	+13	-51	-7	-63	Seriously attacked
"	5	7.1	11.3	-41	-55	-88	-67	
Magnesium with foam removal	3¼	7.1	11.3	-48	-60	-57	-88	"
Steel	2	7.4	10.7	-30	-57	-7	-72	Seriously attacked
"	5	7.4	10.6	-16	-47	-16	-62	
Steel with foam removal	2	7.5	10.6	-21	-46	-19	-78	"
Chlorine cell Graphite	2	7.4	7.2	-31	0	-72	-63	Seriously attacked
"	5	7.2	7.0	-31	-20	-78	-56	
Platinum on titanium	2	7.2	8.4	-19	+6	-11	-29	No attack
"	5	7.1	5.6	-37	0	-57	-57	"
Platinum with foam removal	2	7.5	8.5	-23	-6	-2	-50	No attack
Lead dioxide	2	7.4	7.8	-62	+6	-90	-88	No attack
"	5	7.6	8.4	-26	+8	-97	-85	"
Graphite type GALX[a]	2	7.6	6.8	-23	-7	-82	-44	Seriously attacked
	5	7.6	7.0	-31	-23	-86	-56	
Graphite type GALX-58[a]	2	7.6	6.5	+42	0	-73	-38	Seriously attacked
	5	7.6	7.0	+36	-11	-84	-13	

[a] From National Carbon Co.

have as anodes in secondary effluent. This was done with a cell containing a single anode and cathode spaced 1 inch apart. Polarization measurements were made with a saturated calomel probe, and a General Radio electrometer was used for measuring voltage between the probe and the anode. The probe was located in the center of the anode about 1/32 inch from its surface. The electrometer was connected to the anode through a separate lead to prevent errors caused by the flow of current. A plot was then made of the anode-to-probe voltage versus current density. Measurements were first made in 0.003 molar KCl solution and then in secondary effluent. Studies were made on platinum-plated titanium, lead dioxide, graphite, steel, aluminum, and type 430 stainless steel anodes, each working with a platinum-plated titanium cathode.

The platinum-plated titanium and the lead dioxide anodes behave as noble electrodes. In the tests using these electrodes, very little current was passed until the decomposition potential of the KCl solution was reached. The measured potential was about 1.5 volts and was the same for all electrodes. At this potential, hydrogen was liberated at the platinum-plated cathode, and a mixture of oxygen and chlorine at the anode. When secondary effluent was substituted for the KCl solution, the same decomposition potential was noted and the slopes of the current density versus voltage curves were similar, indicating that the dominant reaction taking place in the KCl solution and the effluent was the same. When iron, stainless steel, and aluminum were run, current started to flow at a much lower potential than was noted for the noble anodes, showing that these anodes were being attacked. There were no significant differences in behavior between the KCl solution and the secondary effluent.

The results of the polarization studies are significant in that they show that the decomposition of water and chloride or the solution of metal are the dominant electrode reactions, and that the small amount of organic material present has not had a profound effect on these reactions.

Statistical Series

Two sets of statistically designed experiments were performed to evaluate the effect of pH, current density, and temperature on several electrode systems. All experiments used a volume of 700 milliliters, so that results are directly comparable. The electrode systems were all identical, that is, a single anode was placed between two cathodes. The space between the electrodes was 0.64 centimeter. The area of the electrode face was 58 square

centimeters, so that anode and cathode each had a total area of 116 square centimeters. Figure 3 shows the graphite anode system as a typical example of the setup used.

The pH of the solution was adjusted by the addition of aqueous sodium hydroxide or phosphoric acid. This introduced an error in the total solids determination, but in no case was this over 10 percent.

The first set of 24 experiments used lead dioxide and National Carbon Company-type GALX graphite anodes. Each variable was tested at two levels: pH 6 or 8; temperature, 25 to 30 or 55 to 60° C; and current density, 6 or 30 milliamperes per square centimeter. At the latter current density, the time of electrolysis was 1 hour; at the former current density, the time was 5 hours. Thus the same quantity of electricity, 12,600 coulombs, was passed through each sample. This ratio of electrical energy to solution volume, 18,000 coulombs per liter, was used in other experiments. A second set of 12 experiments was made with a platinum-plated titanium anode. In this series initial pH was 5 or 8; current densities and temperatures were the same as in the first set. Table 5 shows initial conditions and results for each experiment in both series.

An analysis of the results from the first series shows several significant effects in spite of the scattering of the data. Current density, initial pH, and temperature had little effect on the performance of lead dioxide. In some runs chloride was completely removed. COD was often reduced from 60 to 70 ppm to 5 to 10 ppm, while ABS was reduced from 8 ppm to as low as 1 ppm. Graphite was attacked under all test conditions, but more severely at elevated temperatures and current densities. The anode flaked off solid particles and colored the solution from brown to black. This color was not removed by filtering with Whatman No. 5 paper. No further work was done with the graphite anodes because of this attack.

Analysis of results from the second series of experiments with platinum-plated titanium anodes showed no significant effect of changes in current density or temperature. The initial pH had a small effect on total dissolved solids. At an initial pH of 8, total solids were decreased by about 20 percent and final pH was increased. At an initial pH of 5, total solids increased and final pH was decreased. It is possible that acid electrolysis resulted in oxidation of the organics present to higher molecular weight compounds, such as conversion of alcohols to the corresponding organic acids. This could account for the increase in total solids. If, in alkaline solution, the oxidation proceeds further and

partially degrades the compound, the observed decrease in total solids would occur. No precipitates were observed during any of these electrolyses.

The data from the statistical studies on lead dioxide and platinum-plated titanium anodes were combined and analyzed. The lead dioxide system gave the best reductions of COD, ABS, and chlorides, regardless of initial conditions of pH, current density, and temperature. In no case was it satisfactory for the removal of solids. Platinum-plated titanium was effective in removing solids only in alkaline solution and this averaged around 20 percent. In general, the independent variables caused, at best, minor effects with the exception of the pH effect on the platinum anode.

Systems Using Iron or Iron Alloy Anodes

The early electrolytic systems reported in the literature used iron or plain steel electrodes almost without exception. This was probably due mainly to the economics involved, but in some cases the formation of hydrated iron oxides in the waste water was considered desirable. The most widely tested system, however, that of C. P. Landreth, specifically treated the waste water with lime to raise the pH to about 12, where steel will not dissolve during electrolysis. The lime in this system served a twofold purpose. Not only did it prevent the anodes from dissolving, but it was also beneficial in the treatment of the waste water. As stated above, many critics felt that the lime was more effective than the electrolysis. Recent work done by S. G. Grigoropoulos of the Missouri School of Mines demonstrated the removal of ABS from a synthetic waste solution at a pH of about 7 by use of iron electrodes at a current density of 2 milliamperes per square centimeter.

In order to determine conditions under which iron anodes could be used to treat waste water without being attacked, secondary effluent was electrolyzed by using iron electrodes at a pH of 8.4, 11.0, and 12.7. The results of these tests are given in Table 6. At a pH of 11 or lower, iron anodes erode with relatively high current efficiency. The COD, TS, and ABS are reduced. At least part of this reduction, however, is caused by materials being removed by the voluminous precipitate of $Fe(OH)_3$ formed. In a separate test, ferric chloride was added to secondary effluent and the pH adjusted so as to precipitate $Fe(OH)_3$. This treatment removed 57 percent of the ABS with no electrolysis. It was also found that 50 percent of the ABS could be removed from secondary effluent by adjusting its pH to about 9 and filtering the precipitated calcium phosphate.

1		30	55	5	3.6	-1.4	57	53	-3	59	24	-35	470	644	+174	7	4	-3
2		6	25	8	8.2	+0.2	57	58	+1	59	38	-21	470	348	-122	7	4	-3
3		30	25	8	9.4	+1.4	57	51	-6	59	37	-22	470	372	-98	7	4	-3
4		6	25	5	3.4	-1.6	57	54	-3	59	41	-18	470	643	+164	7	4	-3
5		6	25	8	8.5	+0.5	57	54	-3	59	41	-18	470	360	-110	7	5	-2
6		6	55	5	2.9	-2.1	57	59	+2	59	31	-28	470	630	+160	7	2	-5
7		30	25	5	3.4	-1.6	57	51	-6	59	37	-22	470	620	+150	7	3	-4
8		30	55	8	9.1	+1.1	57	50	-7	59	38	-21	470	410	-60	7	4	-3
9		30	25	8	8.4	+0.4	57	51	-6	59	36	-23	470	380	-90	7	3	-4
10		6	55	8	8.4	+0.4	57	54	-3	59	28	-31	470	330	-140	7	3	-4
11		6	25	5	3.3	-1.7	58	57	-1	56	42	-14	460	630	+170	6	3	-3
12		30	55	5	3.0	-2.0	58	57	-1	56	46	-10	460	590	+130	6	4	-2

[a] Pb = PbO_2, G = Graphite, National Carbon Company, Type GALX.

Table 5. RESULTS, FIRST FACTORIAL SERIES

Run No.	Anode mat[a]	CD, ma/cm^2	T, °C	pH, start	pH, end	ΔpH,	Cl-, start, ppm	Cl-, end ppm	Cl-, Δ, ppm	COD, start, ppm	COD, end, ppm	COD, Δ, ppm	TS, start ppm	TS, end, ppm	TS, Δ, ppm	ABS, start, ppm	ABS, end ppm	ABS, Δ, ppm	Color
1	Pb	6	25	8	---	-----	70	10	-60	48	12	-36	570	530	-40	7	---	---	H_2O
2	G	6	55	8	3.3	-4.7	70	44	-26	48	163	+115	570	555	-15	7	2	-5	Coffee
3	G	6	25	6	3.8	-2.2	70	23	-47	48	37	-11	570	569	-1	7	2	-5	Yellow
4	Pb	30	25	6	6.2	+0.2	71	23	-48	45	13	-32	560	541	-19	7	2	-5	H_2O
5	Pb	6	55	6	4.1	-1.9	71	11	-60	45	11	-34	560	707	+147	7	1	-6+	H_2O
6	G	30	55	6	4.2	-1.8	72	41	-31	50	126	+76	566	684	+118	7	4	-3	Black
7	G	30	25	8	6.7	-1.3	72	6	-66	50	24	-26	566	571	+5	7	4	-3	Yellow
8	Pb	30	55	8	8.2	+0.2	72	12	-60	50	0	-50	566	602	+36	7	1	-6	H_2O
9	G	30	55	8	6.8	-1.2	72	47	-25	57	109	+52	490	550	+60	9	5	-4	Tea
10	Pb	6	25	6	3.6	-2.4	72	15	-57	57	22	-35	490	578	+88	9	1	-8	H_2O
11	G	30	25	6	6.1	+0.1	72	11	-61	57	32	-25	490	582	+92	9	5	-4	Yellow
12	Pb	30	25	8	7.6	-0.4	66	9	-58	57	9	-48	491	507	+16	8	2	-6	H_2O
13	Pb	6	55	8	4.8	-3.2	66	1	-65+	57	5	-52	491	457	+66	8	1	-7+	H_2O
14	Pb	30	55	6	3.1	-2.9	66	8	-58	57	8	-49	491	568	+77	8	1	-7+	H_2O
15	G	6	25	8	6.0	-2.0	66	32	-34	57	47	-10	491	447	-44	8	3	-5	Yellow
16	G	6	55	6	2.8	-3.2	57	47	-10	59	217	+155	470	73	-397	7	4	-3	Black
17	Pb	30	55	8	7.1	-0.9	66	9	-57	57	7	-50	491	540	+49	8	1	-7	H_2O
18	G	30	25	8	7.0	-1.0	66	9	-57	57	31	-26	491	500	+9	8	5	-3	H_2O
19	G	30	55	6	3.8	-2.2	88	70	-18	67	74	+7	630	980	+350	8	6	-2	Yellow
20	G	6	25	6	3.2	-2.8	88	47	-41	67	40	-27	630	840	+210	8	4	-4	Yellow
21	Pb	30	25	6	3.2	-2.8	88	11	-77	67	7	-60	630	480	-150	8	1	-7+	H_2O
22	G	6	55	8	3.0	-5.0	88	47	-41	67	290	+223	630	660	+30	8	4	-4	Black
23	Pb	6	25	8	3.7	-4.3	88	4	-84	67	9	-58	630	400	-230	8	1	-7+	H_2O
24	Pb	6	55	6	3.5	-1.5	88	3	-85	67	4	-63	630	500	-130	8	1	-7+	H_2O

At a pH of around 12, iron becomes passive and does not dissolve when made the anode in a waste-water solution. Under these conditions the iron anode is effective in reducing COD and ABS. It is not, however, as effective as a lead dioxide anode working at a pH of 7. No further work was done on the iron anode system.

Table 6. IRON ANODES IN SECONDARY EFFLUENT

Run	pH	Change, %				Remarks
		COD	TS	Cl	ABS	
1	8.4	-73	-56	-51	-88	20 hr @ 0.1 amp anode eroded at a current efficiency of 91%. Heavy iron floc.
2	11.0	-63	-32	-18	-72	16 hr @ 0.1 amp anode eroded at current efficiency of 100%. Iron and phosphate ppt present.
3	12.7	-53	+370[a]	+5	-72	16 hr @ 0.1 amp anode did not erode. Solution saturated with $Ca(OH)_2$. Heavy phosphate ppt.

[a] Increase in TS due to addition of $Ca(OH)_2$.

Bacteria Kill Studies

The following experiment was performed to confirm the reported effectiveness of electrolytic action in killing bacteria. Raw secondary effluent was electrolyzed by using a volume of 1.5 liters in a glass cell with circulation. The electrodes were platinum-plated titanium, 3 x 3 inches. Five cathodes and four anodes were used at a total current of 2 amperes. Samples of electrolyte were taken at 0, 15, 45, and 180 minutes, and all samples were refrigerated until bacterial counts could be made. All counts were started within 4 hours after the samples were taken. The results are summarized in Table 7. Electrolysis does substantially reduce the bacteria count. Even after 14,400 coulombs have passed, however, some bacteria survive.

Table 7. BACTERIAL COUNTS IN ELECTROLYTICALLY TREATED WASTE WATER

Samples	Coulombs/liter	Standard plate count/ml
1. Original	0	1,280,000
2. After 15 min.	1,200	3,600
3. After 45 min.	3,600	1,200
4. After 180 min.	14,400	240

ESTIMATION OF COLIFORM GROUP DENSITY[a]

Samples	Five fermentation tubes			MPN per 100 ml	Limits of MPN	
	10 ml	1 ml	0.1 ml		Lower	Upper
	PRESUMPTIVE TEST (LB)					
1. Original	5/5[b]	5/5	5/5	Over 1,610		
2. After 15 min.	5/5	5/5	4/5	1,610	390	4,240
3. After 45 min.	5/5	5/5	2/5	542	180	1,405
4. After 180 min.	5/5	5/5	0/5	240	68	754
	CONFIRMED TEST (BGLB)					
1. Original	5/5	5/5	5/5	Over 1,610		
2. After 15 min.	4/5	2/5	0/5	22	7	67
3. After 45 min.	1/5	0/5	0/5	2	0.5	7
4. After 180 min.	0/5	0/5	0/5	0	0	5.9

[a] MPN = most probable number of coliform group organisms.
LB = lactose broth
BGLB = brilliant green lactose broth.

[b] The number in the numerator represents positive tubes, and that in the denominator, the total tubes planted.

Effect of Cl^- Ion on ABS and COD

Tests indicated that the chloride ion in secondary effluent was being converted to hypochlorite during electrolysis and this was probably having an effect on the ABS present. To verify this, a series of experiments was made with synthetic solutions of ABS and KCl whose concentrations were similar to those in secondary effluent. In one series, an indifferent salt, K_2SO_4, was used instead of KCl. These solutions were then electrolyzed with a lead dioxide anode. Table 8 is a summary of results. It will be noted that the presence of chloride is not a necessary condition for removal of ABS with the lead dioxide anode.

Table 8. ELECTROLYSIS OF ABS SOLUTIONS

ELECTROLYSIS OF SOLUTION CONTAINING 55 ppm Cl^- (as KCl) AND 10 ppm ABS		
Coulombs passed	Anode material	Residual ABS, ppm
360	PbO_2	6
1,575	PbO_2	5

ELECTROLYSIS OF SOLUTION CONTAINING 55 ppm K_2SO_4 AND 10 ppm ABS		
Coulombs passed	Anode material	Residual ABS, ppm
360	PbO_2	9
720	PbO_2	7
1,440	PbO_2	6
360	Pt	9
720	Pt	9
1,440	Pt	9

Samples of secondary effluent were then treated with hypochlorite but not electrolyzed. If the hypochlorite alone is destroying the ABS, these samples should show a reduction. Two sets of samples were tested. The first was treated with an

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excess based on the amount of hypochlorite that would be generated if only hypochlorite were being formed at the anode. This is 3,800 ppm hypochlorite. Two samples were treated: In the first a pH of 8 was maintained, and in the second, pH 5. Phosphoric acid was used in both to maintain the pH. The ABS was reduced from 8 to 6 ppm; total solids and chloride were greatly increased. COD could not be determined because of the large chloride concentration.

The second set was treated with a more realistic amount of hypochlorite, 100 ppm. The starting pH was 7.5, but, after addition of the hypochlorite, 8.2. This had no effect whatever on ABS or COD originally present. Total solids and chloride were somewhat increased. Both sets were stirred for 1 hour after addition of the hypochlorite and stored for a minimum of 24 hours before analysis. It is apparent that the hypochlorite alone had little effect on either COD or ABS.

A sample of secondary effluent was treated with chlorine. The gas was intermittently bubbled through the solution to maintain sufficient free chlorine to give a positive test with starch-potassium iodide paper for 1 hour. The solution was stored for 24 hours and analyzed. Total solids and chloride were increased. The ABS was reduced 16 percent and the COD was increased 30 percent. Neither a hypochlorite nor a chlorine treatment behaved as the electrolytic treatment of secondary effluent did.

Relation Between ABS and COD

ABS is the only organic constituent of secondary effluent that can be easily measured quantitatively. An attempt was made to determine the relationship between ABS and COD in the course of an electrolysis. A solution containing 20 ppm ABS was electrolyzed for 12 hours with a lead dioxide anode at a current of 0.2 ampere. Samples were withdrawn periodically and analyzed for ABS and COD. Because of the low current the reaction went very slowly. No correlation between ABS and COD changes was obtained. The measured ABS content decreased only slightly but COD actually increased.

The small decrease in ABS, from 20 to 15 ppm in 12 hours' electrolysis, is worthy of note since the amount and stability of the foam formed by the samples decreased rapidly. For example, after 7 hours' electrolysis, properties of this material were markedly less than those of the starting solution. Since ABS analysis includes unsubstituted benzenesulfonates or sulfonates with shorter chain substitutents, it appears that the electrolysis

effects scission of the alkyl chain, leaving residues of lower surface activity.

Complete Chemical Analysis of Treated and Untreated Secondary Effluent

Total solids, residue at 600° C, available chlorine, chloride, COD, ABS, sulfate, calcium, phosphate, heavy metals, silica, and nitrogen were determined on two 5-liter samples of treated, unpasteurized secondary effluent and on one sample of untreated material. The electrolysis was done in a glass jar by using 293 square centimeters of lead dioxide as anode and an equal area of iron as cathode. (Both electrodes have been used in successive electrolyses with no cleaning or treatment of the electrodes between runs.) The electrolysis was carried out for 205 minutes at a current of 7.3 amperes. The results of this treatment along with those for untreated effluent are shown in Table 9. The reduction in COD and ABS is typical of these electrolytic treatments. (COD is often reduced further, especially when platinum cathodes are used; however, this varies considerably.)

Power Requirements

A treatment of 18,000 coulombs per liter was chosen as the constant dose for the statistical study because it was found that the anodes under study all produced substantial effects on most of the variables at this dose. Although there were wide variations in individual runs, the lead dioxide system in some cases produced reductions of 80 percent in COD, and 90 and 88 percent, respectively, in Cl and ABS concentrations. Since this is a substantial amount of power and would be a major part of the cost of the treatment, it might be expedient to sacrifice some of the effect, in order to lower cost, by reducing the amount of power used.

To study this possibility, a series of tests was run in which the time of treatment was varied. The standard three-electrode cell with a volume of 700 milliliters of secondary effluent, lead dioxide anodes, and platinum-plated cathodes was used. A second series was run by the same procedure, but the foam was removed as formed. These results are summarized in Table 10, and in Figure 4 the results without foam removal are plotted as a function of coulombs passed.

The smaller reduction in COD for those runs in the foam-skimming series is difficult to explain. When the foam is allowed

to remain on the surface, it dries and forms a scum. This scum is then removed in the final filtering before analysis. It may be that some reduction in COD is caused by materials' being carried out of the solution by this scum. The dried scum was never analyzed, but analysis of the liquid formed when the skimmed foam from a 5-liter electrolysis broke gave these concentrations (in ppm): COD 355, ABS 60, TS 330, and chloride 83. This was a sixfold concentration of material in the foam, but the amount carried out by the foam was trivial.

Table 9. COMPLETE ANALYSIS FOR TREATED AND UNTREATED SECONDARY EFFLUENT, ppm

	Treated		Untreated
	Sample I	Sample II	
Residue at 103° C [a]	390	450	490
Weight loss in residue at 600°C	134	140	65
Available chlorine [b]	<1	<1	<1
Chloride	5	9	66
COD	29	30	60
Sulfate (as SO_4)	63	78	73
Calcium	21	14	35
Phosphate (as P_2O_5)	3	6	28
Heavy metals (as lead)	<1	<1	<1
Silica [c]	3	4	0.6
Ammonia nitrogen (as N)	3	12	16
Total nitrogen (less NO_3)	6	13	20
Nitrate (as N)	12	11	11
ABS	2	3	10

[a] Residues comprise dissolved nonvolatile substances plus suspended solids that pass No. 5 filter paper. Some insoluble material is undoubtedly lost from the electrolyzed effluent owing to (a) slight deposits on electrolysis vessel and (perhaps) on electrodes, and (b) traces retained by filter paper.

[b] Although samples are refrigerated pending analysis, there may be some loss of available chlorine following electrolysis.

[c] The small increase in silica is attributed to conducting the electrolysis in a glass vessel.

Because the effect of foam skimming is not clear from these tests, it was not assumed in the cost estimates. The cost was

estimated at two levels of power: 9,000 coulombs per liter where the expected reductions (in percent) are COD 65, ABS 50, and Cl 65; and 18,000 coulombs per liter where the expected reductions are COD 80, ABS 90, and Cl 90.

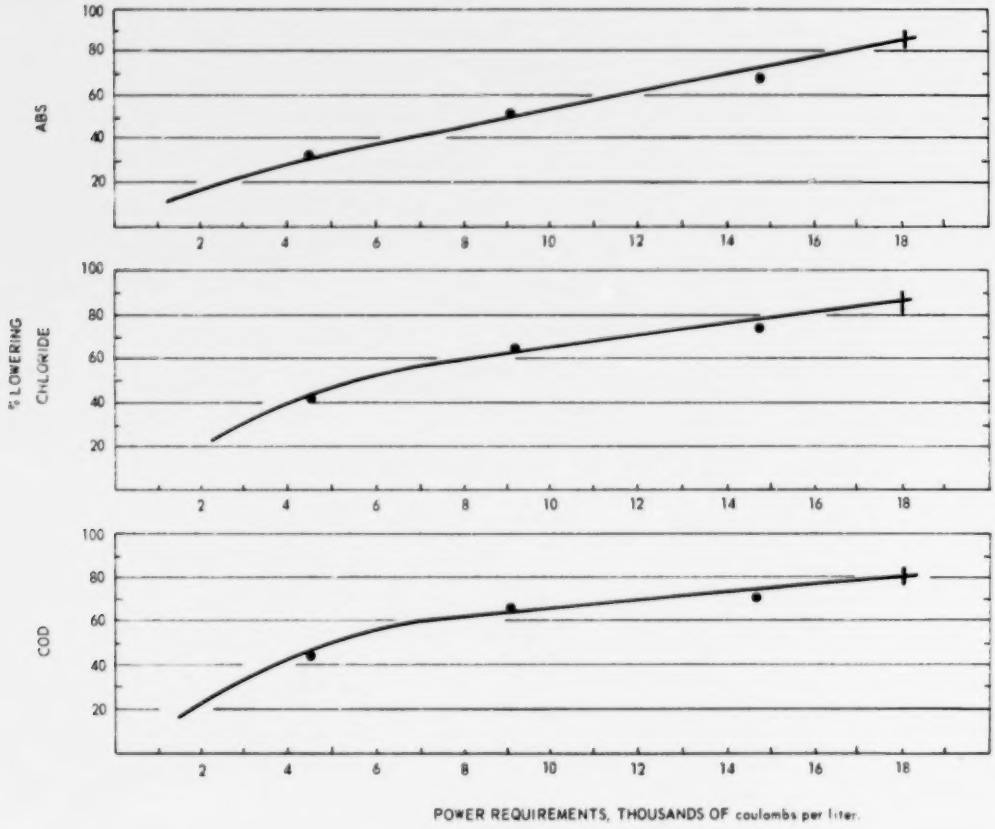

Figure 4. Effect of treatment time on COD, chlorides, and ABS.

Cost of Electrolytic Waste Treatment

Laboratory tests have shown that the best that can be expected from an electrolytic treatment of secondary effluent is a reduction (in percent) of about 80 in COD, 90 in chloride, and 90 in ABS. There will be little effect on total solids. This reduction will be achieved by a dose of 68×10^6 coulombs per 1,000 gallons. At one-half of this dose, or 34×10^6 coulombs, the reductions (in percent) should be COD 65, ABS 50, and chloride 65. The cost of this treatment is largely determined by the amount of electrical power used and the life of the anodes.

An estimate of the cost of treating waste water at two levels of conductivity was made to illustrate the large effect of conductivity on the cost of electrolytic waste treatment. These two levels were 5×10^{-4} and 12×10^{-4} mho per centimeter. The

Table 10. TIME SERIES

Time in min.	CD, ma/cm^2	Coulomb dose per liter	Change, %				Remarks
			COD	Cl^-	TS	ABS	
300	6	18,000	-75	-90	-9	-88	Foam not removed
60	30	18,000	-80	-78	-11	-80	"
45	30	13,500	-70	-72	+26	-66	"
30	30	9,000	-68	-65	+21	-50	"
15	30	4,500	-50	-43	+24	-33	"
60	30	18,000	-48	-92	+8	-95	Foam removed
45	30	13,500	-50	-75	+8	-95	"
30	30	9,000	-46	-62	+8	-94	"
15	30	4,500	-40	-45	+3	-84	"
60	6	3,600	-32	-38	+3	-75	"
45	6	2,700	-30	-28	0	-50	"
15	6	900	0	-8	+8	-50	"
16 hr	0.86	8,200	-54	-59	-45	-30	Foam not removed

lower level represents "fresh effluent" and is the typical secondary effluent tested in this study. The higher level, called brackish effluent, would be for a solution with about 1,000 ppm salt.

Both the power consumed and the number of anodes required are dependent on the anode current density. Since the statistical study showed that the performance of the cell was not affected by current density, it is possible to choose an operation point that will minimize the cost without affecting the treatment. This operation point was chosen by plotting estimated power and anode cost versus current density. The following assumptions were used in these estimates:

1. Coulombs/1,000 gal — 68×10^6

2. Cell voltage:

Current density, ma/cm^2	Volts: Fresh effluent	Volts: Brackish effluent*
1	3.3	3.2
6	7.8	5.5
9		6.9
30	27.9	14.9

3. Lead dioxide anode size — 1-1/4 x 6 x 30 in

4. Active anode area — 2 ft^2/anode

5. Anode cost — $20.00 each

6. Anode life — 1,000 days**

7. Effluent conductivity, mho/cm at 25°C,
   - Fresh effluent — 5×10^{-4}
   - Brackish effluent* — 12×10^{-4}

8. Electrical power — $0.005/kw-hr

The minimum power and anode cost for the treatment of secondary effluent with a conductivity of 5×10^{-4} mho/cm and an anode life of 1,000 days is achieved by operating at a current density of about 6 ma/cm^2. The current density for effluent with a conductivity equal to brackish effluent would be about 9 ma/cm^2.

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* With 1,000 ppm NaCl.

** Estimated from experience with lead dioxide anodes in other applications.

For the cost estimate, a basic unit was taken, consisting of four electrolytic tanks operated in series from a single power supply. For fresh effluent, each unit will contain 72,000 1-1/4- x 6- x 30-inch anodes mounted between steel sheets as cathodes. This unit will be capable of treating 1 mgd at the 68×10^6 coulombs per 1,000 gallons dose for the maximum effect, or 2 mgd for the somewhat lower treatment. For the brackish effluent, with the same power unit as for the fresh waste, 60,000 1-1/4- x 6- x 30-inch anodes would be required, and this unit could treat 1.25 mgd at the maximum dose or 2.5 mgd at the lower dose. Plants for 10 and 100 mgd are made by grouping 5 or 50 of these units for the low-level or 10 and 100 for the high-level fresh-effluent treatment. For the brackish effluent where the basic unit is 1.25 mgd, the groupings are 4 or 40 for the low-level and 8 or 80 for the high-level treatment.

Not included in the estimates are a sand filter for the discharge, site preparation, roads, landscaping, fences, land, area lighting, laboratories or other buildings, and the cost of any polishing operations needed to make the product potable. A summary of the cost is given in Table 11. A more detailed breakdown is given in Appendix 3.

Table 11. ELECTROLYTIC TREATMENT COST FOR FRESH AND BRACKISH EFFLUENT, $

	Fresh effluent		Brackish effluent	
	Low dose	High dose	Low dose	High dose
Cost per 1,000 gal	1.21	2.42	0.90	1.80
Plant costs				
2 mgd	2,700,000	5,260,000	1,906,000[a]	3,701,000[a]
2.5 mgd[c]			2,383,000	4,626,000
10 mgd	12,940,000	25,740,000	9,112,000	18,084,000
100 mgd	128,140,000	256,140,000	89,860,000	179,580,000

[a] The calculated cost of a 2.5 mgd plant was normalized to 2 mgd by multiplying by 0.8.

Incomplete Information

During the work the following items were not completely investigated because of lack of time. Information on these subjects would be helpful in the complete evaluation of the electrolytic processes taking place. It was felt, however, that, in view of the high cost of the treatment, further work to evaluate these items was not justified:

1. The amount of chloride converted to chlorate. Although some effort was spent in developing a method to determine the chlorate content of the electrolyzed effluent, time did not permit a full evaluation of this procedure.

2. The effect of cathode material in the electrolytic treatment. Only two cathode materials were used in this study, iron and platinum-plated titanium. There may be some evidence that the platinum cathode was a little more effective in reducing the COD than the iron was. Lead is also a possible material.

3. Lead-silver alloys. Lead-silver alloys are reported to form insoluble lead dioxide coats when made anodic in chloride solution. In one trial in this study this behavior was not noted. A more complete investigation might find conditions where these alloys could be used. If so, a considerable saving in anode costs should be possible. The Lead Industries Association study will certainly look into this possiblity.

4. Foam removal. It was observed during this study that foam formed electrolytically is denser than foam formed mechanically. This may be caused by rapid diffusion of hydrogen gas through the bubble wall, which allows it to shrink without breaking, or by the production of smaller bubbles formed electrolytically rather than mechanically. In the laboratory test there appeared to be about a sixfold concentration of impurities in the electrolylically formed foam. The volume of this foam was such, however, that, in the one analysis made, this did not represent a significant removal of impurities from the main body of the liquid. There was considerable scatter of data for foam removal, which made its evaluation impossible.

5. Analysis of effluent and foam. Analyses of treated and nontreated secondary effluent are shown in Table 9. The ignition of residue at 600° C shows that treatment diminishes ash but increases the mass of combustible material.

Our only interpretation is that organic materials, for example, polysaccharides, gain weight during electrolysis by addition of oxygen or replacement of hydrogen by oxygen or chlorine. The remarkable reduction in chloride content has been noted in all analyzed runs. A minor amount of chloride may go to chlorate (not included in these values) and some may go to form organic chlorine compounds. We believe most chlorine is lost as such (or as chlorine oxides) in the evolved oxygen and hydrogen. The gases have a strong chlorine odor and react with iodide paper. No quantitative estimate has, however, been made.

The large loss in phosphate was unexpected. It may have been removed with the scum formed from the collapsed foam. Foam formed during electrolysis of a separate 5-liter run was removed for study. It collapsed to about 100 milliliters of liquid, whose analysis is shown in Table 12.

Table 12. ANALYSIS OF ELECTROLYTIC FOAM

	Found in foam, ppm	Change in treated effluent if foam were returned, ppm
TS	330	6
ABS	60	1
COD	355	7
Chloride	83	1.5

APPENDIX A.

A SURVEY
OF THE PUBLISHED WORK
ON ELECTROLYTIC TREATMENT
OF WASTE WATER

APPENDIX A.
A SURVEY OF THE PUBLISHED WORK ON ELECTROLYTIC TREATMENT OF WASTE WATER

This survey covers both patent and technical literature in the field of electrolytic treatment of municipal and industrial wastes. Municipal waste is defined as that material originating in homes and the like consisting primarily of human excretions. Industrial waste is material arising from industrial processes. It is recognized that industrial wastes are often dumped into the sanitary waste system and the resultant mixture processed in the local waste treatment plant.

The survey was made by searching Chemical Abstracts, Industrial Arts Index, and the patent art. The original article was read for all pertinent references unless otherwise noted. All methods reported are described in detail below.

The earliest record found of electrolytic treatment of waste is a patent [1] issued to William Webster in 1889. The waste water electrolyzed with iron electrodes at a potential of about 10 volts. If insufficient chlorides were present to generate chlorine, sea water or salt was added. Iron electrodes were specified to take advantage of the formation of hydrated iron oxides, which aided in sludge flocculation. A plant was built at Crossness, a suburb of London, England, but apparently nothing further was done.

In 1909, J. T. Harris received a patent [2] on a method for the purification of waste water by electrolysis. This sytem is unique in that both iron and aluminum are used together as anodes and allowed to corrode during electrolysis. It was claimed that if either metal is used alone it cannot be completely precipitated; however, almost complete removal of metal ions can be achieved with the combination, and better clarification of the solution was obtained. Copper is employed to aid in sterilization. A small electrode, either anode or cathode, is claimed to be sufficient and does not add harmful amounts of copper to the solution. This multiple metal system is of interest as a means of introducing metal hydroxides to aid in sludge flocculation.

In 1913, G. W. Swinburne received a patent [3] for a method of purifying waste water by electrolysis. The patent did not specify a particular type of apparatus but required that a chloride be added and the solution be electrolyzed. No fixed amount of chloride was called for, only that sufficient amount be added to give the "degree of purification desired." Apparently, a system was never set up, since no further mention is made in the literature.

Although electrolytic waste treatment plants were in operation as early as 1911 in Santa Monica, California, and Oklahoma City, Oklahoma, much of the technical literature and about half of the patents concerning electrolytic waste-water treatment in the United States are related to the system patented [4] and promoted by C. P. Landreth. The apparatus consisted of a rectangular wooden box containing many vertical steel plates about 1/2 inch apart and so arranged that the waste water flowed in thin sheets among them. The plates were alternately connected to the positive and negative terminals of a DC power supply. The exact number of electrodes varied, but one demonstration unit built at Elmhurst, New York City, contained 1,100 electrodes 10 by 16 by 3/16 inches. The spaces among the plates contained paddles mounted on shafts passing through and insulated from the electrodes. The paddles were rotated to assure thorough mixing of the solutions, to prevent polarization of the electrodes, and to remove any scum formed on the electrodes. Electrodes were made of steel, but other metals could be used. The power required was determined by the conductivity of the waste water and the condition of the electrodes, since with use the required power increased. The waste water was dosed with lime just prior to entering the electrolyzer. This served to increase the conductivity of the waste water and raise the pH, which passivated the steel, making it erode more slowly, and also aided in settling of solids after electrolysis. Residence time was about 1 minute but varied from unit to unit depending on size. The effluent from the electrolyzer was run into settling tanks and then discharged. It was described as clear, odorless, and nonputrescent. (Present laboratory work has shown electrolyzed secondary effluent from an activated-sludge plant to be odorless and often perfectly clear. It is well known that very good bacteria kills can be obtained by electrolysis. See page 22 of this report for a bacteriological study carried out in this laboratory.)

The active promotion of electrolytic waste-water treatment by Landreth aroused considerable controversy. As stated above, electrolytic plants had been in operation as early as 1911 and good results had been reported. The system was claimed to be ideally suited for built-up areas because of the relatively small land requirements and the absence of odor compared with conventional plants. Demonstration plants were built in the Elmhurst section of New York City, and in Allentown and Easton, Pennsylvania, among others. [5]

All these plants were praised at first for their supposedly high-quality effluent and lack of odor. Some reservations were made on operating cost since sludge from the settling tanks had to be hauled away and this increased the overall cost. Compared

with present-day activated-sludge plants, this might not be an objection. In time, however, all the plants were abandoned. It was claimed that the electrolysis did nothing that could not be achieved by dosing with lime at the rate prescribed for electrolysis. The data given in reports on the several plants are not in agreement on the efficiency of this method. The oxygen demand and total solids varied, as would be expected, from plant to plant. In most cases the total solids increased, probably from the addition of large amounts of lime. The result of the controversy was that all electrolytic treatment plants had been abandoned by about 1930. No doubt the single biggest factor was the cost of operation, which was estimated to be as much as double that of more conventional methods.[6]

In 1930 an electrolytic system of removing grease from waste water was patented.[7] It consisted of a concrete conduit 6 feet deep by 9 feet wide and 150 feet long. There were two sets of electrodes, spaced about 100 feet apart. The anodes were closely spaced graphite rods and the cathodes were steel plates. The anode and cathode were 2 inches apart and the steel plate cathodes abutted alternate walls. The throughput of the system was 7 mgd. Since the electrodes were so closely spaced, the velocity was very high in the electrode area. It was stated that the grease in the waste water coalesced into a hard, odorless layer in the areas between electrodes. Despite this apparently successful operation, no further report of this process can be found.

A report[8] was made on preliminary studies carried out in the Soviet Union on an electrolytic method of treating waste water. Experiments were done on a laboratory scale with iron and graphite electrodes. Various combinations of single- and multiple-electrode systems with and without diaphragms were tested. The main work was carried out on stationary systems. Since this study was made primarily to explore the possibility of substituting electrolysis for chlorination in waste-water treatment, the main effort was directed towards bacteria kill. Satisfactory results were obtained in a diaphragm cell with waste water in the anode compartment and water in the cathode compartment. It was stated that the method was to be scaled up, but no further report can be found in the literature.

Some work has been done on industrial waste treatment. C. J. Brockman has received two patents[9] for the electrolytic treatment of "whole slop" from alcohol fermentation. These patents are primarily concerned with the coagulation and recovery of solids in the slop. An electrolytic process for the removal of phenolic material, thiosulfates, thiocyanates, and chlorides has

been described.[10] No specification of the cell is made other than for electrode material. Platinum mesh anode and iron mesh cathode are preferred.

Two methods for the removal of cyanides from plating wastes have been patented. The first[11] involves DC electrolysis at 93° C and fairly high current densities. It is claimed that concentrated solutions of cyanides can be reduced to several ppm in a period varying from several hours to several days depending on concentration and composition of the solution. Electrode material is not specified. The second method[12] varies somewhat from the above in that the electrolysis is carried out at ambient temperature. Moreover, the anode is constructed of porous carbon, and air or oxygen diffusing through the carbon assists in oxidation of the cyanide. It is possible that a system such as this could be of use in the electrolytic treatment of waste water.

Recently, work has begun in Europe on the electrolysis of waste water mixed with sea water. The object is to use compounds present in the sea water; hypochlorite is generated, and the magnesium salts present form precipitates that aid in sludge flocculation. Not much detail is given[13] on the system currently in use in Norway, but a cost of 6 cents per 1,000 gallons of waste water treated is claimed with a residence time of less than 1 hour and with power cost at $0.01 per kilowatt-hour. In a pilot unit at the University of Naples,[14] horizontal electrodes without diaphragms are used. The Naples unit uses about one-third sea water and, with a residence time of 10 seconds or less, reduces the BOD to about two-thirds of its initial value. Work is apparently still under way on the Italian system, and a patent[15] has just been issued for the Norwegian method.

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APPENDIX B.

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APPENDIX B.

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APPENDIX C.

COST ESTIMATES

APPENDIX C.

COST ESTIMATES

The cost estimates include: Cost estimates for fresh-effluent electrolytic plants of various capacities; processing costs for fresh effluent; cost estimates for brackish-effluent electrolytic plants of various capacities; processing costs for brackish effluent; Figure 5 - proposed design of the fresh-water electrolytic plant; Figure 6 - proposed design of a brackish-water electrolytic plant.

The following assumptions were used in these estimates.

1. Coulombs/1,000 gal

   Low dose 34×10^6

2. Cell voltage:

Current density, ma/cm^2	Fresh effluent, volts	Brackish effluent (1,000 ppm NaCl), volts
1	3.3	3.2
6	7.8	5.5
9	---	6.9
30	27.9	14.9

3. Lead dioxide anode size 1-1/4 x 6 x 30 in.

4. Active anode area $2\ ft^2$/anode

5. Anode cost $20.00 each

6. Anode life 1,000 days

7. Effluent conductivity

   mhos/cm at 25° C

Fresh effluent	5×10^{-4}
Brackish effluent (1,000 ppm NaCl)	12×10^{-4}

Notes on estimates:

1. The most economical plant to build and operate for fresh waters was the one with a current density of 6 ma/cm^2 across the anodes. For the brackish waters, the plant with 9 ma/cm^2 across the anodes was the most economical. Our studies are based on these two plants.

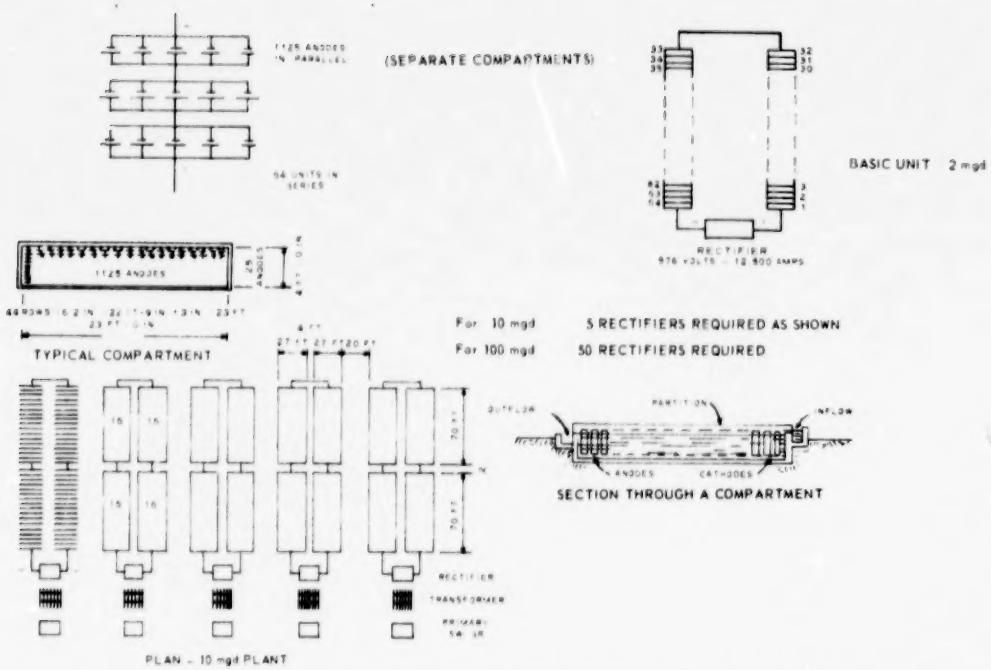
(SEPARATE COMPARTMENTS)
BASIC UNIT 2 mgd
TYPICAL COMPARTMENT
For 10 mgd 5 RECTIFIERS REQUIRED AS SHOWN
For 100 mgd 50 RECTIFIERS REQUIRED
SECTION THROUGH A COMPARTMENT
PLAN - 10 mgd PLANT

Figure 5. Proposed design of a fresh-water electrolytic plant (current density 6 ma/cm^2).

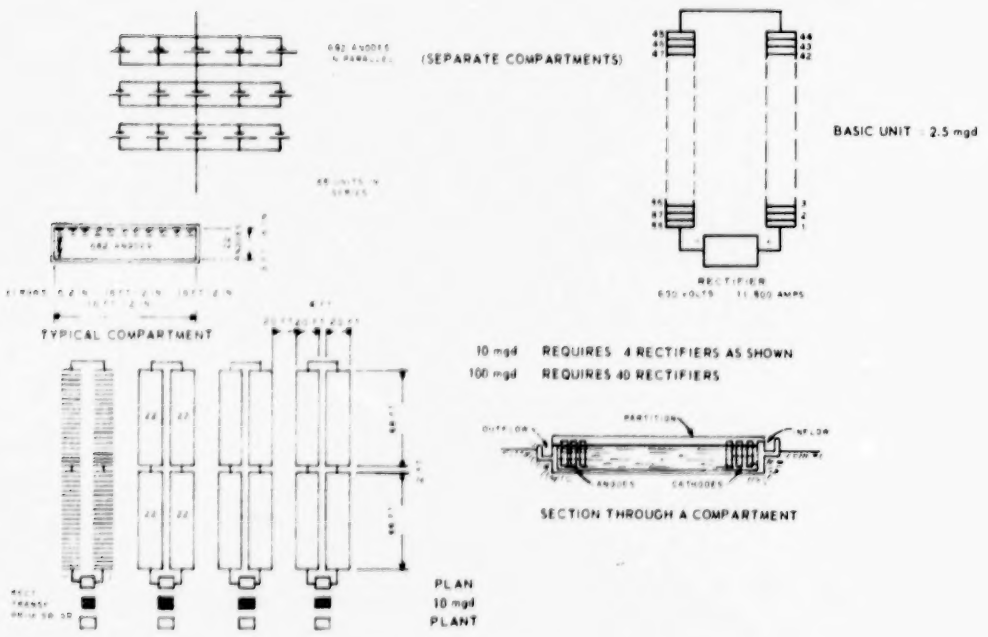

Figure 6. Proposed design of a brackish-water electrolytic plant (current density 9 ma/cm^2).

2. The battery limits for water flow are the feeding trenches along the concrete cell tanks and the discharging trenches on the opposite side of the same tanks. Others must provide the necessary sewers to get the waters to and from these points.

3. The electrification begins with the high-voltage primary switch gear.

4. Not included in the estimates are site preparations, roads, landscaping, fences, land, area lighting, and laboratories or other buildings.

5. A traveling powered-maintenance bridge on set of ground rails is included with each basic plant.

6. For the processing costs, 34×10^6 coulombs per 1,000 gallons was used in computing the power consumption figures.

PROCESSING COSTS, $: FRESH-WATER TREATMENT (6 ma/cm^2)

Operating month	30 days	
Monthly treatment	60 million gal	
Power	4,404,000 kw-hr x $0.005 per kw-hr	= 22,020
Labor	Operators, 288 mhr/month @ $3.00/mhr	= 864
Maintenance		
Anode replacement, * 2,160 x ($20.00 + $0.50)		= 45,000
Cathode replacement, 437 x ($0.70 + $0.50)		= 530
Depreciation		
Plant investment, $2,700,000 less cathode-anode investment of $1,490,400 = $1,209,600		
$1,209,600 on a 25-year life		= 4,000/month
Total processing costs per month		= 72,414
Processing costs per 1,000 gallons		
$72,414 ÷ 60,000 = $1.21		

If power cost is $0.01 per kw-hr, total processing cost per month would be $94,434 and the cost per 1,000 gallons would be $1.57.

* Anode replacement based on $\frac{30}{1000}$ of 72,000 for every 30 days at cost of $20.00 each plus $0.50 installation charge.

Table 13. COST ESTIMATE: FRESH-WATER ELECTROLYTIC PLANT

(CURRENT DENSITY, 6 ma/cm², CAPACITY, 2 mgd)

Items, $	Material	Labor	Total
Excavations		2,400	2,400
Foundations (electrical gear)	4,000	3,000	7,000
Electrolytic tanks (4)	16,000	16,000	32,000
Tank membranes[a]	3,000	9,000	12,000
Anodes (72,000)	1,440,000	18,000	1,458,000
Cathodes (72,000)	50,400	18,000	68,400
Anode-cathode supports	36,000	18,000	54,000
Primary sw gr, transformers, rectifier, DC sw, metering, alarms (7,200 kw)	180,000	36,000	216,000
Busses (main)	35,000	30,000	65,000
Anode-cathode connectors	36,000	18,000	54,000
Traveling maintenance bridge	15,000	10,000	25,000
Subtotals	1,815,400	178,400	1,993,800
Contingencies	370,000	36,000	406,000
Total labor and material	2,185,400	214,400	2,399,800
Overheads			
Engineering (not repetitive)		150,000	150,000
Contractor's profit and overheads		150,000	150,000
Supervision		30,000	
Construction services and equipment		55,000	
Insurances and bonds		30,000	
Fee		35,000	
Total cost of project			2,699,800
Say			2,700,000
Plant costs			
2 mgd	2,700,000		
10 mgd	12,940,000		
$2,700,000 + 4 (2,560,000)[b]			
100 mgd	128,140,000		
$2,700,000 + 49 (2,560,000)[b]			

[a] These membranes are used in the construction of the cement cell tanks to make them waterproof and electrically insulated.

[b] Since the major part of engineering would be involved in the initial plant, these costs would not be repeated in the plants that followed. If $10,000 is allowed for engineering, additional plants would cost $2,560,000 each.

TABLE 14. COST ESTIMATE: BRACKISH-WATER ELECTROLYTIC PLANT

(CURRENT DENSITY, 9 ma/cm², CAPACITY, 2.5 mgd)

Item, $	Material	Labor	Total
Excavations		2,400	2,400
Foundations (electrical gear)	4,000	3,000	7,000
Electrolytic tanks (4)	17,200	17,200	34,400
Tank membranes[a]	2,400	7,000	9,400
Anodes (60,000)	1,200,000	15,000	1,215,000
Cathodes (60,000)	42,000	15,000	57,000
Anode-cathode supports	30,000	15,000	45,000
Primary sw gr, transformer, rectifier, DC sw, metering, alarms (6,900 kw)	175,000	35,000	210,000
Busses (main)	42,000	36,000	78,000
Anode-cathode connectors	30,000	15,000	45,000
Traveling maintenance bridge	15,000	10,000	25,000
Subtotals	1,557,600	170,600	1,728,200
Contingencies	320,000	34,000	354,000
Total labor and maintenance	1,877,600	204,600	2,082,200
Overheads			
Engineering (not repetitive)		150,000	150,000
Contractor's profit and overheads			150,000
Supervision		30,000	
Construction services and equipment		55,000	
Insurance and bonds		30,000	
Fee		35,000	
Total cost of project			2,382,200
Say			2,383,000

Plant costs	
2.5 mgd	2,383,000
10 mgd	9,112,000
2,383,000 + 3 (2,243,000)[b]	
100 mgd	89,860,000
2,383,000 + 39 (2,243,000)[b]	

[a] These membranes are used in the construction of the cement cell tanks to make them waterproof and electrically insulated.

[b] Since the major part of engineering would be involved in the initial plant, these costs would not be repeated in the plants that followed. If $10,000 is allowed for engineering, additional plants would cost $2,243,000 each.

PROCESSING COSTS, $: BRACKISH-WATER TREATMENT ($9\ ma/cm^2$)

Operating month	30 days	
Monthly treatment	75 million gal	
Power	4,920,000 kw-hr x $0.005 per kw-hr	= 24,600
Labor	Operators, 288 mhr/month at $3.00/mhr	= 864
Maintenance		
Anode replacement	1,825 x ($20 + $0.50)	= 37,415
Cathode replacement	365 x ($0.70 + $0.50)	= 438
Depreciation		
Plant investment, $2,383,000 less cathode-anode investment of $1,242,000 = $1,141,000		
$1,141,000 on a 25-year life		= 3,800/month
Total processing costs per month		= 67,117
Processing costs per 1,000 gal		
$67,117 ÷ 75,000 = $0.895		

If the power costs $0.01 per kw-hr, total processing cost per month would be $91,717 and the cost per 1,000 gallons would be $1.22.

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(Continued from inside front cover.)

		Report Number
AWTR- 8	Ultimate Disposal of Advanced-Treatment Waste	999-WP-10
	Part 1. Injection	
	Part 2. Placement in Underground Cavities	
	Part 3. Spreading	
AWTR- 9	Adsorption of Biochemically Resistant Materials from Solution. I	999-WP-11
AWTR-10	Feasibility of Granular, Activated-Carbon Adsorption for Waste-Water Renovation	999-WP-12
AWTR-11	Evaluation of the Use of Activated Carbon and Chemical Regenerants in Treatment of Waste Water	999-WP-13
AWTR-12	Evaluation of Various Adsorbents and Coagulants for Waste-Water Renovation	999-WP-14
AWTR-13	Electrochemical Treatment of Municipal Waste Water	999-WP-19